IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Hiroya OKUMURA et al.

Serial No. 09/950,081 Group Art Unit 1745

Filed September 12, 2001

Examiner Mr./Mrs. Mark RUTHKOSKY

For: SEPARATOR FOR SOLID POLYMER TYPE FUEL CELL AND PROCESS FOR PRODUCING THE SAME

DECLARATION UNDER RULE 132

Honorable Commissioner of Patent and Trademarks,
Washington, D.C.

Sir,

I, Koji TAKABATAKE, declare:

That I am a citizen of Japan, residing at 3-6-19-311 Nyoidani, Minoo-shi, Osaka, Japan;

That I was born on December 2, 1949 in Okayama and graduated from the Department of Synthetic Chemistry, Faculty of Engineering Science, Osaka University, Toyonaka, Japan, 560 in March 1972;

That I have been employed by Nippon Shokubai Kagaku Kogyo Co., Ltd. (current NIPPON SHOKUBAI Co., Ltd.), JAPAN since 1972, and engaged in the research in thermosetting resins and sheet, and bulk molding

compounds;

That I have been on loan to Japan Composite Co., LTD. which is a joint venture company of Mitsui Takeda Chemicals, Inc. and NIPPON SHOKUBAI Co., Ltd. since April 2003, and engaged in the research in separators for fuel cell, which was taken over from Mitsui Takeda Chemicals, Inc., as Senior Chief Researcher since April 2004; and

That the following experiments were conducted under my direct supervision;

EXPERIMENT

I investigated effectiveness of pressurized kneading on compounds and shaped articles thereof.

(Preparation of resin composition)

To a four-neck flask equipped with a stirrer, a condenser, a nitrogen-inlet, and thermometer were charged 374 g of bisphenol A-type epoxy resin (manufactured by Toto Kasei Co. Ltd., epototoYD128, epoxy equivalent 187 g/eq), 172 g of methacrylic acid, 0.2 g of triphenylphosphine, 0.1 g of hydroquinone as a thermal polymerization inhibitor, and reacted for 8 hours at 120 °C to obtain 546 g of vinyl ester resin having an acid value of 1.8 mgKOH/g. The vinyl ester resin was diluted with 364 g of styrene monomer to obtain a resin composition.

(Preparation of molded plate)

Sample A

This sample corresponds to Example 5 of the present specification.

The resin composition (224 g) was kneaded with 28 g of styrene-butadiene block copolymer (D-KX410CS, Shell JSR Elastomer), 330 g of artificial graphite powder (manufactured by SEC Co. Ltd., SGL10, average particle size of 10 μm), 770 g of artificial graphite powder (manufactured by SEC Co. Ltd., SGL25, average particle size of 25 μm) and 5.6 g of t-butyl peroxybenzoate (manufactured by Nippon Yushi Co. Ltd., TBPB) by a kneader (manufactured by Toshin Co., Ltd., THM0.5-3 type hybrid mixer, orifice of 10 cm x 9 cm, depth of 15 cm). In this kneading step, these powdery components could not be sufficiently commingled, and only a coarse particle was formed. It was impossible to make the mixture uniform. Further, it took about 2 hours to make the mixture coarse particulate. Since this manner was too time-consuming, each component was reduced to one-half in amount and subjected to the kneading step. As a result, it took about 13 minutes to make the kneaded compound coarse particulate. The compound was further kneaded for 2 minutes. That is, the total kneading time was 15 minutes.

After deairing the kneaded compound, the resulting

matter was cured in a plate mold (300 x 300 x 8 mm) under 100 kg/cm^2 (9.8 MPa) at 150°C for 2 minutes to obtain a molded plate.

The surface of the resulting molded plate (300 x 300 x 2 mm) was colored black with white stripped patterns all over the surface thereof. Moreover, while the central region of the plate was blackish, the corner part thereof was matte and grayish. It seems that nonuniformity of the molded plate is attributable to the state of the kneaded compound, that is, inhomogeneous coarse particulate.

Sample B

This sample corresponds to Example 6 of the present specification.

A kneaded compound was prepared in similar manner to Sample A except for kneading with the use of a pressure kneader under a pressure of 3.92 x 10^6 Pa (4 kgf/cm^2), and at 40°C and 50 rpm. In this kneading step, these powdery components were sufficiently kneaded to give a clay-like or viscous matter. Further, it took about 3 minutes to make the mixture clay-like. In order to unify the kneading condition (kneading time) to Sample A, the mixture was further kneaded for 12 minutes so that the total kneading time was 15 minutes.

The kneaded compound was molded to the same manner as Sample A to give a molded plate.

The resulting molded plate (300 x 300 x 2 mm) was homogeneous, and the surface thereof was uniformly colored black.

Sample C

This sample is based on Component 23648 described in Table 8C of US Patent No. 6,251,308. In Component 23648, the vinyl ester resin used is taken to be the same as that of Samples A and B mentioned above. In this sample, the same resin composition, graphite and additive as those of Samples A and B were used. Moreover, in accordance with Component 23648, polyisocyanate and calcium stearate were added as modifier and mold release agent to the system. Incidentally, since details of polyisocyanate in Component 23648 was unknown, diphenylmethane-4,4'-diisocyanate was used as the polyisocyanate and the amount thereof was adjusted so that the content of the NCO group relative to the vinyl ester resin corresponded to that of Component 23648.

A kneaded compound was obtained in the similar manner to Sample A except for using 11.8 g of calcium stearate (manufactured by Nippon Yushi Co. Ltd., GF200) and 22.6 g of diphenylmethane-4,4'-diisocyanate (manufactured by Mitsui Takeda Chemicals, Inc., Cosmonate PH; NCO content of 33.6%) as modifiers in addition to the kneading components of Sample A. In the kneading step,

these powdery components could not be sufficiently kneaded, and only a coarse particle was formed. It was impossible to make the mixture uniform. Further, it took about 1 hour to make the mixture coarse particulate. Since this manner was too time-consuming, each component was reduced to one-half in amount and subjected to the kneading step. As a result, it took about 10 minutes to make the kneaded compound coarse particulate. In order to unify the kneading condition to Sample A, the mixture was further kneaded for 5 minutes so that the total kneading time was 15 minutes.

The resulting molded plate (300 x 300 x 2 mm) was black with white stripped patterns all over the surface of the plate. Moreover, while the central region of the plate was blackish, the corner part thereof was matte and grayish. It seems that nonuniformity of the molded plate is attributable to the state of the kneaded compound, that is, inhomogeneous coarse particulate.

In the kneaded compounds and the plates obtained in Samples A to C, kneadability, external appearance, and uniformity were determined according to the following methods.

(Kneadability)

In the compositions used in Samples A to C,

kneadability was evaluated based on visual observation of the resulting molded plate and the time required for kneading.

(External appearance)

The surface state of the plate was visually observed and evaluated based on the following criteria.

A: a surface of a plate is entirely uniform.

B: there are white stripped patterns all over a surface of a blackish plate. Moreover, while the central region of the plate was blackish, the corner part thereof was matte and grayish.

The results are shown in Table A.

Table A

		Samples		
		1	2	3
Resin composition [g]		224	224	224
Styrene-butadiene block copolymer [g]		28	28	28
Graphite powder	SGL10 [g]	330	330	330
	SGL25 [g]	770	770	770
t-Butyl peroxybenzoate [g]		2.2	2.2	2.2
Modifier	Calcium stearate [g]	0	0	11.8
	Diphenylmethane-4,4'-diisocyanate [g]	0	0	22.6
Kneadability	Compound state	coarse particulate	clay-like or viscous	coarse particulate
	Required time	2 hrs.	3 min.	1 hrs.
External appearance		B	A	B

EVALUATION

As apparent from Table A, the molded article obtained in Sample B possesses higher reliability than that in Sample A or C. In addition, it is apparent that the pressurized kneading improves the kneading efficiency of the mixture and results in good productivity.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 23rd day of February, 2005

Koji Takabatake

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